

# A Reappraisal of the Electric Dipole Moment Enhancement Factor for Thallium

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The electric dipole moment (EDM) enhancement factor of atomic Tl is of considerable interest as it has been used in determining the most accurate intrinsic electron EDM limit to date. However, the value of this quantity varies from  $-179$  to  $-1041$  in different approximations. In view of the large uncertainties associated with many of these calculations, we have employed the relativistic coupled-cluster theory with single and double excitations and a subset of leading triple excitations and obtained the EDM enhancement factor of Tl as  $-466$ , which in combination with the most accurate measured value of Tl EDM yields  $2.0 \times 10^{-27} \text{ e cm}$  as the new upper limit for the EDM of the electron. The importance of all-order correlation effects is emphasized and their trends are compared with those of two other ab initio calculations.

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The EDM of a nondegenerate physical system [1, 2] arises from the simultaneous violations of parity ( $\mathcal{P}$ ) and time-reversal ( $\mathcal{T}$ ) symmetries. The invariance of  $\mathcal{T}$ -symmetry is also associated with the invariance of the  $\mathcal{CP}$  (combined charge conjugation ( $\mathcal{C}$ ) and  $\mathcal{P}$  symmetries) symmetry on the basis of the  $\mathcal{CPT}$  theorem [3]. Thus, EDMs of atoms can shed light on  $\mathcal{CP}$  violations originating in the leptonic, semi-leptonic and hadronic sectors [4, 5], some of which are not well understood so far. The knowledge of EDMs provide valuable insights into some profound questions such as the existence of new physics beyond the standard model (SM) and the matter-antimatter asymmetry in the universe [4–6]. Given the current interest in understanding different types of  $\mathcal{CP}$  violations originating from elementary particles both by accelerator and non-accelerator based approaches, EDM searches are of great significance.

The EDMs of paramagnetic atoms are sensitive to the the EDM of the electron [7]. The most accurate limit for the latter has been obtained by combining the results of the Tl EDM measurement and the EDM enhancement factor (EF), defined as the ratio of the EDM of the atom to that of the electron, of this atom. Liu and Kelly have calculated this EF, and found its value to be  $-585$ , with an error bar of  $5 - 10\%$ , using a linearized version of the relativistic coupled-cluster (RCC) theory [8]. A more recent calculation of this quantity for Tl by Dzuba and Flambaum uses a hybrid approach combining the configuration interaction (CI) method and many-body perturbation theory (MBPT) and reports  $-582$  with an estimated accuracy of  $3\%$  [9]. Both these results, coincidentally, are in good agreement with each other. Nevertheless, considering the fact that the values of the Tl EDM EF reported in the literature range from  $-179$  to  $-1041$  [13–17], there is clearly a need for high precision calculations of this quantity. Thus, the primary focus of

this paper is to determine the EF of Tl by proceeding beyond [8, 9]. An accurate treatment of the unusually strong electron correlation effects in the ground state Tl EF warrants the use of an all-order relativistic many-body method like the RCC theory.

The open-shell RCC theory with single, double and a subset of leading triple excitations employed in the calculation of the EDM EFs is discussed in detail in [10–12]. However, we briefly present below the salient features of this method for the sake of completeness. The effective one electron form of the interaction Hamiltonian due to the electron EDM is given by [12],

$$H_{EDM}^{eff} = 2ic \sum_j \beta_j \gamma_j^5 \vec{p}_j^2 \quad (1)$$

where  $\beta$  and  $\gamma^5$  are the usual Dirac matrices,  $\vec{p}_j$  is the momentum vector of the  $j^{th}$  electron. As the strength of the EDM interaction is sufficiently weak, we consider the wavefunction expansion only up to first-order in perturbation. Thus, the modified atomic wavefunction for a valence electron ‘ $v$ ’ is given by,

$$|\Psi'_v\rangle = |\Psi_v^{(0)}\rangle + \left(\frac{d_e}{ea_0}\right) |\Psi_v^{(1)}\rangle. \quad (2)$$

In the RCC theory, the unperturbed and perturbed wavefunctions can be expressed as [10–12],

$$|\Psi_v^{(0)}\rangle = e^{T^{(0)}} \{1 + S_v^{(0)}\} |\Phi_v\rangle \quad (3)$$

$$\text{and } |\Psi_v^{(1)}\rangle = e^{T^{(0)}} \{T^{(1)} (1 + S_v^{(0)}) + S_v^{(1)}\} |\Phi_v\rangle \quad (4)$$

where  $|\Phi_v\rangle$  is the Dirac-Fock (DF) wavefunction obtained by appending the valence electron  $v$  to the closed-shell ( $[5d^{10}6s^2]$ ) reference wavefunction,  $T^{(0)}$  and  $S_v^{(0)}$  are the excitation operators for core and valence electrons in an

unperturbed case, where as,  $T^{(1)}$  and  $S_v^{(1)}$  are their first order corrections. The atomic wavefunctions are calculated using the Dirac-Coulomb Hamiltonian given by,

$$H_0 = \sum_i \{c\alpha_i \cdot p_i + (\beta_i - 1)m_i c^2 + V_n(r_i)\} + \sum_{i < j} V_C(r_{ij}), \quad (5)$$

where  $\alpha$  and  $\beta$  are Dirac matrices,  $V_n(r_i)$  is the nuclear potential and  $V_C(r_{ij})$  is the Coulomb potential on the electron  $i$  due to the  $j^{th}$  electron.

We consider only the single and double excitation operators in the expansion of the RCC wavefunctions (termed as CCSD approximation), by defining,

$$T = T_1 + T_2 \quad \text{and} \quad S_v = S_{1v} + S_{2v}, \quad (6)$$

for both the perturbed and unperturbed operators. Further, we construct triple excitation operators for  $S_v^{(0)}$  as,

$$S_{vab}^{pqr,(0)} = \frac{\widehat{H_0 T_2^{(0)}} + \widehat{H_0 S_{2v}^{(0)}}}{\epsilon_v + \epsilon_a + \epsilon_b - \epsilon_p - \epsilon_q - \epsilon_r}, \quad (7)$$

which are used to evaluate CCSD amplitudes iteratively. This is referred to as CCSD(T) method. Here,  $\epsilon_i$  is the single particle energy of an orbital  $i$ .

The final expression for the EDM EF ( $\mathcal{R} = \frac{D_a}{d_e}$ ) in terms of the coupled-cluster operators is given by,

$$\mathcal{R} = \frac{1}{ea_0} \frac{\langle \Phi_v | \{1 + S_v^{(0)\dagger}\} \overline{D^{(0)}} \{T^{(1)}(1 + S_v^{(0)}) + S_v^{(1)}\} | \Phi_v \rangle}{\langle \Phi_v | e^{T^{(0)\dagger}} e^{T^{(0)}} + S_v^{(0)\dagger} e^{T^{(0)\dagger}} e^{T^{(0)}} S_v^{(0)} | \Phi_v \rangle} + \text{h.c.} \quad (8)$$

where the dressed operator  $\overline{D^{(0)}} = e^{T^{(0)\dagger}} \vec{D} e^{T^{(0)}}$  and  $\vec{D} = e \vec{r}$  is the electric dipole moment operator due to the applied electric field. The procedure for the calculation of the above expression is discussed elsewhere [10–12].

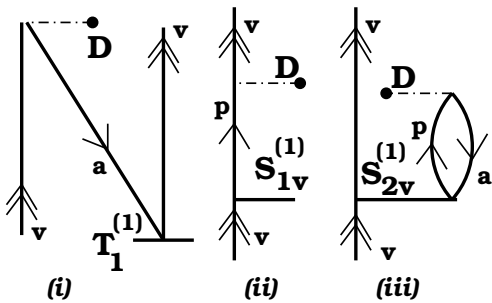


FIG. 1: The leading correlation diagrams: (i)  $D T_1^{(1)}$ , (ii)  $D S_{1v}^{(1)}$ , (iii)  $D S_{2v}^{(1)}$ . The exchange and hermitian conjugate diagrams are not shown. Labels  $v$ ,  $a$  and  $p$  refer to valence, core and virtual orbitals, respectively.

In Table I, we present the contributions from different RCC terms along with the lowest order (DF) contributions to  $\mathcal{R}$  and compare them with the results reported

TABLE I: The contributions from various RCC terms to the EDM EF ( $\mathcal{R}$ ) of Tl. A new quantity,  $X = T + S_v$  is defined to compare our results with those of [8].

Term	This Work	Liu & Kelly [8]
$(D T_1^{(1)})_{\text{lowest-order}}$	−153.6	−153.2
$(D S_{1v}^{(1)})_{\text{lowest-order}}$	−268.5	−267.3
$(D T_1^{(1)})_{\text{higher-order}}$	−224.7	−342.1
$(D S_{1v}^{(1)})_{\text{higher-order}}$	−45.5	−102.5
$D X_2^{(1)}$	248.0	240.9
$X_1^{(0)\dagger} D X_1^{(1)}$	22.5	22.4
$X_2^{(0)\dagger} D X_1^{(1)}$	−78.2	49.3
$X_2^{(0)\dagger} D X_2^{(1)}$	21.5	−36.9
$X_2^{(0)\dagger} D X_1^{(1)} X_2^{(0)}$	−4.3	−2.2
Higher order RCC terms	13.0	—
Normalization contribution	3.8	6.5
Total EDM EF	−466	−585

by Liu and Kelly [8]. It is evident from this table that the bulk of the contributions to  $\mathcal{R}$  comes from the RCC terms  $D T_1^{(1)}$ ,  $D S_{1v}^{(1)}$  and  $D S_{2v}^{(1)}$ . These important all-order correlation effects involving the core, valence and core-valence sectors are shown diagrammatically in Fig.1. The single largest contribution (−378), to  $\mathcal{R}$  comes from the all-order core correlation effect of which its lowest order contribution is less than half of its magnitude. The all-order valence correlation contribution from  $D S_{1v}^{(1)}$  is −314 while its DF contribution is −269. The difference between the all-order and the lowest-order contributions of the terms  $D S_{1v}^{(1)}$  and  $D T_1^{(1)}$  clearly indicate that the all-order core correlations are of crucial importance in the case of Tl. It is indeed significant that one of the classes of the all-order core-polarization effects represented by  $D S_{2v}^{(1)}$  makes the largest positive contribution, of magnitude 248. This dramatically reduces the final result. There is a non-negligible contribution from the other higher order terms; however, many of them cancel each other to give an effective value of only about 13. The normalization of the RCC wavefunction also gives a contribution of 4 and the total result for the ground state EDM enhancement factor for Tl due to the intrinsic EDM of the electron amounts to −466.

The  $H_{EDM}^{eff}$  being an odd-parity operator mixes the atomic states of opposite parities, however, with the same angular momentum. Therefore, we have investigated the role of various intermediate states of  $s$  symmetry. The RCC terms such as:  $D T_1^{(1)}$ ,  $D S_{1v}^{(1)}$ ,  $S_{1v}^{(0)\dagger} D S_{1v}^{(1)}$  and  $S_{2v}^{(0)\dagger} D S_{1v}^{(1)}$ , have got significant contributions from the orbitals, from 5s through 11s and are shown in Fig. 2. For comparison, we have also shown the DF contributions from these individual orbitals. The correlation effects for the 6s orbital are much larger than its DF contribution

itself. The core electrons of  $d$  symmetry also influence the RCC amplitudes indirectly. High accuracy calculations should therefore employ an all-order method such as the RCC theory to account for the strong correlation effects in a comprehensive manner.

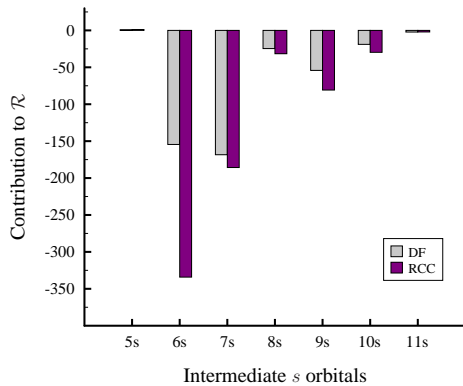


FIG. 2: The contributions from singly excited intermediate states, 5s through 11s, in DF and RCC approximations are compared.

We have also investigated the role of different doubly excited states that contribute through  $DS_{2v}^{(1)}$ . These contributions are given in Table II. As seen from the table, the largest contribution comes when the 6s orbital is excited to the valence orbital,  $6p_{1/2}$  followed by the latter's fine structure partner, the  $6p_{3/2}$  orbital. There are also, however, some non-negligible contributions coming from the 5d orbitals when they get excited to the virtual  $p_{3/2}$  and  $f_{5/2,7/2}$  orbitals.

TABLE II: Contributions from selected doubly excited states through  $DS_{2v}^{(1)}$  term.

core( $a$ )	virtual( $p$ )	Results	core( $a$ )	virtual( $p$ )	Results
6s	$6p_{1/2}$	170.88	5d <sub>3/2</sub>	$8f_{5/2}$	0.92
6s	$7p_{1/2}$	3.61	5d <sub>3/2</sub>	$9f_{5/2}$	2.06
6s	$8p_{1/2}$	2.01	5d <sub>5/2</sub>	$6p_{3/2}$	5.37
6s	$9p_{1/2}$	4.50	5d <sub>5/2</sub>	$7p_{3/2}$	0.76
6s	$6p_{3/2}$	31.42	5d <sub>5/2</sub>	$8p_{3/2}$	0.59
6s	$7p_{3/2}$	3.00	5d <sub>5/2</sub>	$9p_{3/2}$	1.46
6s	$8p_{3/2}$	1.95	5d <sub>5/2</sub>	$8f_{7/2}$	2.34
6s	$9p_{3/2}$	5.26	5d <sub>5/2</sub>	$9f_{7/2}$	5.36

The EDM enhancement factor result for Tl from different calculations are compared in Table III. It is quite apparent that the published results lie in a rather large range. Although, the overall trends of the majority of the correlation terms in [8] appear similar to ours, there is a significant difference in the magnitude of many of those

correlation contributions. This can be due to the several approximations considered in their calculation, some of which are the following: (i) they have considered only the one-body form for the unperturbed atomic Hamiltonian and as well as for the EDM Hamiltonian thereby neglecting the important contributions partly from the DF potential and largely from the two electron Coulomb interaction. (ii) they have considered only the linear terms and a few selected non-linear terms in their calculations. (iii) they have included selective triple excitations approximately in the unperturbed singles amplitude equations only (i.e., in  $T_1^{(0)}$  and  $S_{1v}^{(0)}$  equations), where as, they are completely ignored in the unperturbed doubles equations. (iv) they have frozen the inner core up to the 4s orbital for the calculation of unperturbed amplitudes, where as, for solving the perturbed doubles equations they further freeze 4s, 4p and 4d orbitals. In contrast to the above set backs, we consider all the non-linear terms arising from the single and double excitations. We also consider the leading triple excitations in both the unperturbed singles and doubles cluster equations. We solve both the unperturbed and perturbed, closed- and open-shell equations, self-consistently. We have performed the all-electron relativistic CCSD(T) calculation and the total active orbitals considered here, are: 14s,  $13p_{1/2,3/2}$ ,  $13d_{3/2,5/3}$ ,  $9f_{5/2,7/2}$  and  $8g_{7/2,9/2}$ .

The comparison between the results of Dzuba and Flambaum based on a combined CI+MBPT approach [9] and our all-order CCSD(T) is not straight forward. They have considered the three outer shell electrons ( $6s^2 6p_{1/2}$ ) as valence and the rest as core. The core, virtual and valence orbitals are generated in a  $V^{N-3}$  closed-shell potential in contrast to the  $V^{N-1}$  potential used in our calculation. The valence-valence correlations are evaluated by CI, while the valence-core and the core-core correlations by MBPT. In view of the strong correlation effects in Tl, it is important to treat them by an all-order correlation method instead of dealing with it as a three valence system within the framework of an hybrid CI and finite order MBPT approach. It appears from the previous work of Dzuba and Flambaum that the  $\mathcal{P}$  &  $\mathcal{T}$  violating Hamiltonian used in [9], considers only the internal electric field due to the nucleus and not the electrons. The major drawback of the latter work is that all the correlations obtained by MBPT (except for s-electrons in the one-body correlation operator  $\Sigma_1$  [9]) are considered only up to second order although they have emphasized the importance of valence-core correlations in their paper. In contrast, we have considered all these effects to all-orders in the residual Coulomb interaction in the framework of the full fledged CCSD(T) theory. The CI+MBPT result of [9] compares well with that of [8]. The latter as mentioned earlier is based on a linearized CCSD(T) approach with several approximations. However, the agreement between these two results is fortuitous.

We have also compared our result with a few *ab initio*

TABLE III: Comparison of the ground state EDM EF ( $\mathcal{R}$ ) of Tl from different calculations.

$\mathcal{R}$	Method	Reference
-466(10)	CCSD(T)	This Work
-582(20)	CI+MBPT	Dzuba 2009 [9]
-585(30 - 60)	LCCSD(T)	Liu 1992 [8]
-179	MBPT(2)	Hartley 1990[13]
-301	MBPT(2)	Kraftmakher 1988[14]
-502, -562, -607, -1041	MBPT(1)	Johnson 1986 [15]
-500	semi-emp.	Flambaum 1976 [16]
-716	semi-emp.	Sandars 1975 [17]

finite-order MBPT results [13–15] and semi-empirical estimations [16, 17] in Table III. Apparently, the MBPT calculations [13, 14] have under estimated the results, where as, the first-order MBPT calculation with semi-empirical potentials [15] and the semi-empirical calculations [16, 17] have over estimated the EDM EF of Tl.

The results of a few relevant physical quantities such as the electric dipole (E1) matrix element for  $7S \rightarrow 6P_{1/2}$  transition and the magnetic dipole hyperfine structure constants of the ground state  $6P_{1/2}$  and the lowest singly excited state  $7S$  have been presented in Table IV. It is clear that, our results are in better agreement with the experiments than those reported in [9].

TABLE IV: Comparison of the E1 matrix element and hyperfine structure constants of low-lying states in Tl.

Transition	This Work	Dzuba & Flambaum [9]	Expt.
	E1 tr. amp.		
$7S \rightarrow 6P_{1/2}$	1.82	1.73	1.81(2) [18]
State	Magnetic dipole hfs constant		
$6P_{1/2}$	21053	21067	21311 [19]
$7S$	11894	11417	12297 [20]

The error estimates quoted in [9] for Tl EDM EF does not seem to be convincing for the reason that they have treated the valence-core correlations for the most part only up to second order in perturbation theory. Clearly an all-order correlation calculation is needed for a reliable error estimation. Although, in the CCSD(T) approach the effect of the dominant triple excitations are considered while evaluating the CCSD amplitudes, their direct contributions are ignored while computing Eq. (8). To estimate the maximum possible error on the EF result, we evaluate the contribution coming from the triple excitations, such as,  $S_{vab}^{pqr,(0)}$  and  $S_{vab}^{pqr,(1)}$  where the latter is defined as (for details, see [21]),

$$S_{vab}^{pqr,(1)} = \frac{\widehat{H_0 T_2^{(1)}} + \widehat{H_0 S_{2v}^{(1)}}}{\epsilon_v + \epsilon_a + \epsilon_b - \epsilon_p - \epsilon_q - \epsilon_r}. \quad (9)$$

Considering the error due to the basis set incompleteness and the error due to the neglected triples, we obtain the final result for the EDM EF of Tl to be  $-466(10)$ . By combining the EDM experimental result for Tl,  $-(4.0 \pm 4.3) \times 10^{-25} \text{ e cm}$ , given by Regan et al. [22] and our accurate theoretical EDM EF, we obtain a new limit for the electron EDM to be  $(8.7 \pm 9.3) \times 10^{-28} \text{ e cm}$  which translates into an upper limit,  $d_e < 2.0 \times 10^{-27} \text{ e cm}$  at 90% confidence level. In conclusion, we have obtained the most accurate limit for the electron EDM to date by improving the value of the EDM EF for the ground state of atomic Tl by treating the unusually strong electron correlation effects to all-order using the relativistic CCSD(T) method.

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